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ANNUAL LETTER REPORT

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for the period November 1, 1989 - October 31, 1990

Contract #N00014-88-K-0299

"Spontaneous and Induced Coherent Radiation
Generation in Atomic Vapors"

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Annual Letter Report Contract N00014-88-K-0299
Period October 31 1989 to present

Experiments have been performed to study the nonlinear behavior of intense incoherent light propagating in an optical fiber. ~~We have also~~ ^{to} ~~continued our use of~~ incoherent light to study time delayed four-wave-mixing in semiconductor doped glasses and organic dyes and coherent light to generate attosecond beats in potassium vapor. ~~We have~~ ^{to} calculated the effect of the dynamic Lorentz shift on four-wave parametric interactions in a strongly driven two-level system as well as the distinct problem of the nonlinear intensity dependent frequency shift of the reflectivity from a gas dielectric interface. We are presently considering the problem of induced phase modulation by an incoherent pump ~~and have made~~ a diversion in studying an aspect of optical memories using photon echoes.

I

Broad-Band Time-Delayed Four-Wave-Mixing

Experimental work has continued in the area of time delayed four wave mixing (TDFWM) in organic dyes. The impetus of this work is to reconcile the TDFWM technique with short pulse experiments done elsewhere.¹ We have made improvements in our experimental setup. Our sample is now identical to the one used by Becker et al. Previously, we had dissolved Nile Blue in ethanol and placed it in a 1mm thick sample cell. Now, we are flowing Nile Blue dissolved in ethylene glycol through a .1mm dye jet. This insures that the sample is not hole burned and that any discrepancies in dephasing due to dye concentration or type of solvent are eliminated. Also, we have made improvements in our data acquisition software which take into account fluctuations in the source intensity. This helps us account for a fluctuating signal and also allows us to properly subtract off background generated from the excitation pulses.

II

Spectral Broadening

We have performed experiments which demonstrate that when incoherent light of sufficient intensity propagates through a length of optical fiber, the spectral characteristics of the incoherent light are changed. Using a fifty meter long six micrometer diameter fiber we observe a slight increase in bandwidth at power levels around a few watts. This broadening may be partially attributed to self phase modulation.² A significant amount of stimulated Raman scattering is also

seen at these powers which makes it hard to conclude that self phase modulation is the mechanism for the broadening. We hope to resolve this issue by causing a weak probe to propagate down the fiber simultaneously with the strong pump. If the probe pulse is tuned far to the blue with respect to the pump, then induced phase modulation may occur which will broaden the probe without the complications of stimulated Raman scattering. We are presently investigating this problem.

III

Erasing Photon Echoes

An experiment has been set up which will demonstrate the feasibility of using long lived stimulated photon echoes for erasable optical memory. Many groups have previously shown that the stimulated photon echo is a potential technique for storing and retrieving optical data and it has been shown that storage times as long as five hours are possible.³ In a typical experiment to demonstrate this optical storage a pulse of light is first used to irradiate the storage medium. This is called the reference pulse. Subsequently the optical data to be stored is directed into the storage medium and creates a ground state population grating that may persist for a long time compared to the excited state lifetime due to magnetic degeneracy and/or hyperfine structure in the ground state. By applying an additional light pulse a stimulated echo may be produced from the information remaining in the ground state. The stimulated echo represents the recalled data. The experiment that we are doing in sodium vapor will demonstrate whether it is possible to erase data once it has been stored. This will be achieved by applying a pair of pulses identical to the reference and data pulses except that the relative time delay between these third and fourth pulses is a half an optical period greater than the relative time delay between the reference and data pulses. The ground state grating created by the third and fourth pulse should exactly cancel that created by the reference and data pulse.

IV

Attosecond Beats in Atomic Vapors

We are continuing our study of beating effects in time-delayed four-wave mixing (TDFWM) experiments. In all of our TDFWM experiments, two beams with respective momentum vectors k_1 and k_2 are aligned to cross in the atomic vapor cell. Four-wave mixing signals are then emitted in the $2k_1-k_2$ and the $2k_2-k_1$ directions. Higher-order mixing signals are emitted in the $k_1+n(k_1-k_2)$ and $k_2+n(k_2-k_1)$ directions for $n>1$. In our beat experiments, the four-wave and higher-order mixing signal intensities are

monitored and plotted as a function of the time delay between the beams in the k_1 and k_2 directions.

In an experiment in sodium vapor, we have observed sub-femtosecond modulation of the signal intensity as a function of delay time. The shape of the modulation (for $n=1$) is sinusoidal. Theoretical calculations predict that in the weak-field regime (i.e. for small pump pulse area), the width of the beats in the higher-order mixing signals narrows according to

$I_n \propto \left[\frac{1}{2} + \frac{1}{2} \cos(\Omega\tau) \right]^{2n+1}$. A further prediction is that narrowing of the beats will also occur for pump fields with large pulse areas, which can be attained by increasing the laser power or using longer pulses. The excitation fields contained frequency components corresponding to the two fine-structure split levels of the first excited state. The frequency of the modulation was the sum of the frequencies of these two levels. The same excitation scheme is now being employed in an experiment in potassium vapor. The excitation fields are the second harmonics of the fields from two coherent (laser) sources tuned respectively to the fine-structure split levels of the 4S-6P transition. The sum of these frequencies, and therefore the expected modulation frequency, is 1.7×10^{15} Hz. This implies a beat period of 575 attoseconds (1 asec = 10^{-18} sec) which we believe will be the fastest such quantum beat observed. Furthermore, apparatus improvements will hopefully lead to an experimental check of the high-order narrowing effect, and data acquisition software modifications will aid the study of pulse area dependence.

These beats have not yet been observed in our potassium experiment. The relative delay between the fields in the k_1 and k_2 directions is controlled by moving one mirror in a Michelson-type interferometer with a piezoelectric crystal. Since one complete modulation of the signal occurs for every 575 asec of increasing (or decreasing) delay, all mirrors in the interferometer must be stable to much better than 1/4 of one wavelength ($\lambda = 3447\text{\AA}$). We have been successful in stabilizing the delay jitter to well within the limit required for this experiment. Delay jitter in an earlier setup is believed to have made the higher-order narrowing effect impossible to observe.

At present fluctuations in the four-wave mixing signal have prevented observation of the beats. Initially it was thought that amplitude jitter in the pump beams was the main culprit. Small jitter in our Nd:YAG laser amplitude is magnified by three nonlinear processes: dye laser pumping, 2nd harmonic generation, and four-wave mixing. An attempt was made

to eliminate this source of noise by software-selecting data based on pump laser amplitude. Large four-wave mixing amplitude jitter persisted even with the use of this data acquisition software. Other possible sources of fluctuations are the dye lasers and the sample itself.

We have improved on the previous design of the experiment by replacing our former dye lasers (which fluctuated among several cavity modes) with two single-mode dye lasers. We have succeeded in getting them to operate in a single longitudinal mode for the duration of an experimental run. The sample (potassium in a heat pipe oven) is currently being investigated by measuring absorption as a function of atomic number density and buffer gas pressure. Shortly we will search for the modulating mixing signal using the single-mode lasers.

V

Third Harmonic Generation Response and Three-Photon Frequency Shift on Non-Collinear Excitation

We have calculated the 3-photon resonance absorption shift obtained with two noncollinear excitation beams of arbitrary orientation incident on an ensemble of collisionally broadened two level atoms excited close to $1/3$ the resonance frequency. For counterpropagating beams the absorption resonance is narrow and shifted by the order of the linewidth to the violet. As the beams are angled away from the counterpropagating condition the shift increases and if the angle approaches copropagation the shift becomes very large. These predictions have recently been confirmed.⁴ When the shift exceeds the single-photon absorption linewidth by $> L/\lambda$, the shifted line weakens and disappears, but a separate unshifted peak persists at copropagation. The third harmonic generation response is more complicated. Except in the region where the beams are almost copropagating, the shift in third harmonic generation response follows the shift in the absorption response. In contrast to the absorption response the third harmonic generation response increases as the excitation beams approach copropagation, then peaks, then diminishes, broadens and forms a complicated structure whose character is determined by phase matching. At its peak, the intensity of the third harmonic exceeds that obtained from a single beam by L/λ .

VI

Optimization of Third Harmonic Generation In Gases

We have calculated the three-photon resonantly enhanced third harmonic generation in a collisionally self-broadened gas of two level

atoms, appropriately buffered to allow phase matching and have found that it achieves maximum output when the input frequency is sufficiently detuned from exact resonance to avoid excessive self absorption of the third harmonic. For a given sample length and active gas density there exists a combined frequency detuning and buffer gas density which maximizes the third harmonic intensity. This optimal combination depends only on the physical characteristics of the tripling medium and the buffer gas. For this optimal condition αL is of order unity, where α is the absorption coefficient associated with the third harmonic. The characteristics of three-photon resonantly enhanced third harmonic generation in a buffered gas are analyzed in detail.

VII

Intensity Dependent Spectral Reflectivity of a Dense Gas-Dielectric Interface

Intense light incident on a dense gas-dielectric interface, from the dielectric side, is reflected because of the combined effect of the discontinuities in the index of refraction at the interface and at the transition region in the gas where the degree of atomic saturation changes abruptly. This latter discontinuity gives rise to a self-reflected wave. At low intensities this self-reflected wave is weak, the problem is linear, and the net reflected wave intensity taken as a function of frequency has a simple shape which should allow measurement of the hitherto undetected self-induced frequency shift in a collision-broadened gas at an absorption resonance. Calculations show that in the weak intensity regime, r'' , the imaginary part of the reflection coefficient is quasi-Lorentzian and shifted by an amount comparable to its width. At high light intensity r'' broadens, shifts, and distorts. The distortion, which nonuniformly modulates the Lorentzian shape, comes from an interference of the forward and self-reflected waves. In the wings of the resonance line the self-reflected wave can be neglected, and the shape of r'' though not distorted is shifted from where it would be if there were no LFC (local field correction).

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